

LATERAL DIFFUSION ELECTRODES, PART IV

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LATERAL DIFFUSION ELECTRODES, PART IV

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The use of lateral diffusion electrodes, manufactured by powder metallurgy instead of by fritting, in fuel cells is discussed, with a detailed description of the diffusion processes within the active carbon coating. The physical principles underlying the mass transport of the reactive gas through the microporous and macroporous structure of the active carbon are explained, to substantiate the statement that smaller electrode surfaces, shorter interelectrode spacing, and light-weight current collector grids are possible, resulting in considerable weight reduction of fuel cells (to about 5 kg per kilowatt). Means for further reduction in weight and cost price are listed, including spiral design of the electrodes, low-density materials for casings and other parts, small amounts of catalyst, use of air as oxidizer, reduction in volume of the electrolyte by greater compactness of the electrodes, etc. Future use for spacecraft propulsion is envisioned.

Author

1. Lateral Diffusion

The electrochemical properties of catalysts with an active carbon base are no doubt closely linked to the highly specific structure of carbon. The cata-

* Numbers in the margin indicate pagination in the original foreign text.

lyst coating of the electrodes actually constitutes a twin network of pores. Thus, the macrochannels separating the carbon grains and the microchannels existing within each grain must be taken into consideration.

Various studies have shown that, on immersing the electrode into the electrolyte, the macrochannels are completely invaded by the fluid. Conversely, we are of the opinion - in accordance with the hypothesis formulated by Hillebrand and Iacksonen (Bibl.64 - 65) - that the microchannels remain filled with gas and serve to transport the gaseous reagents to points where the electrochemical reactions take place.

So as to make certain that the gas always remains within the microchannels, it must be assumed that the liquid never enters these channels, either during the treatment by the solution used as chemical deposition of the catalyst or during normal operation of the electrode. We found that, as soon as liquid penetrates into the micropores, it becomes extremely difficult to remove it again and that the diffusion of the gas in the electrode will be blocked. This was demonstrated during an experiment which consisted in polarizing the electrode after stoppage of the gas feed. At this instant, the gaseous reagent contained in the microchannels was consumed so that, because of the decrease in pressure, the liquid was able to invade these channels. This greatly lowered /26 the performance of the electrode, in an irreversible manner.

The fact that the solution of the chemical deposition does not enter the micropores seems to indicate that the catalytic metal is deposited only on the surface of the carbon grains and that, after mixing with the binder, only the macrochannels will present electrolytically active surfaces.

Investigations were made to determine whether the type of bond between the catalytic metal and the carbon might have an influence on the catalytic proper-

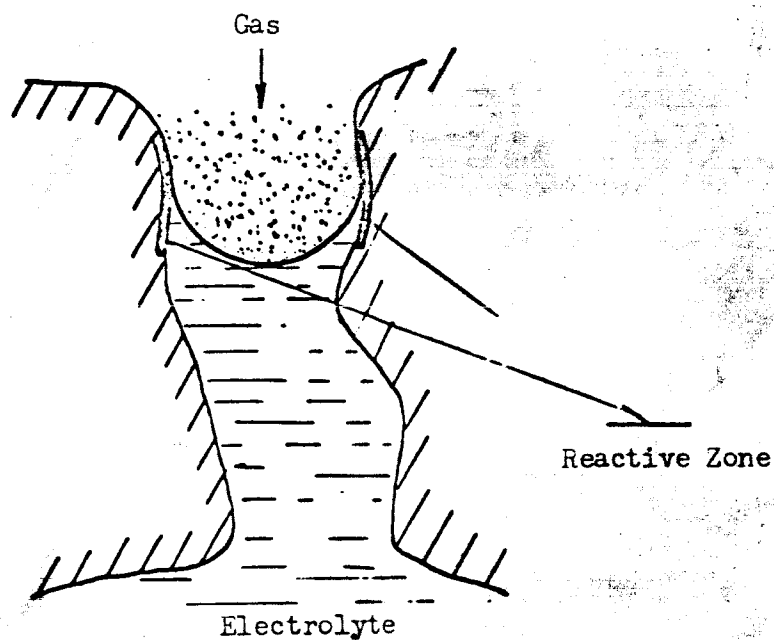


Fig.13a Case of a Classic Porous Material

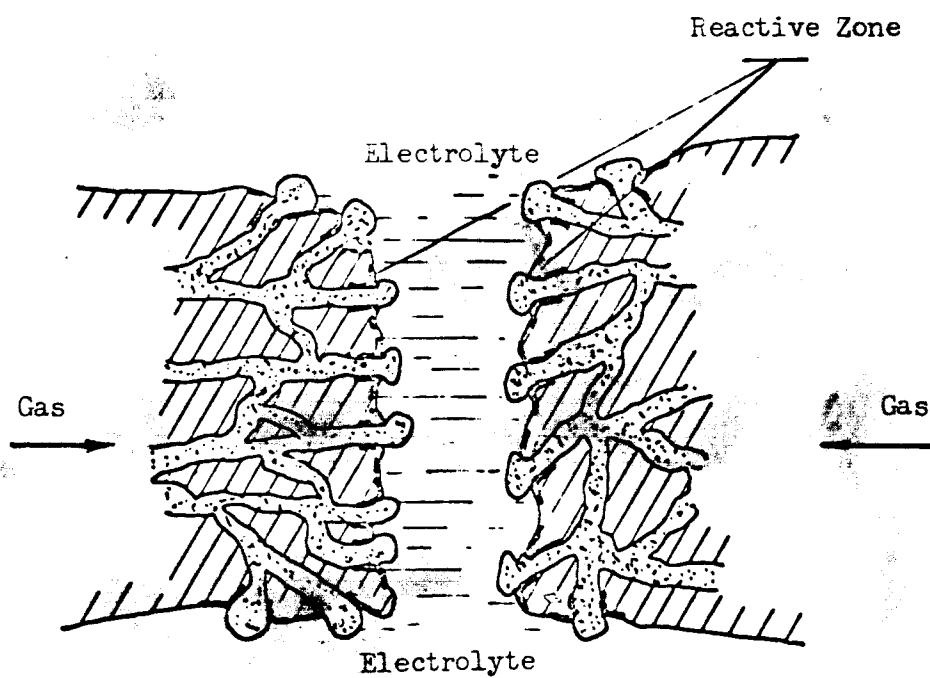


Fig.13b Case of a Lateral Diffusion Electrode

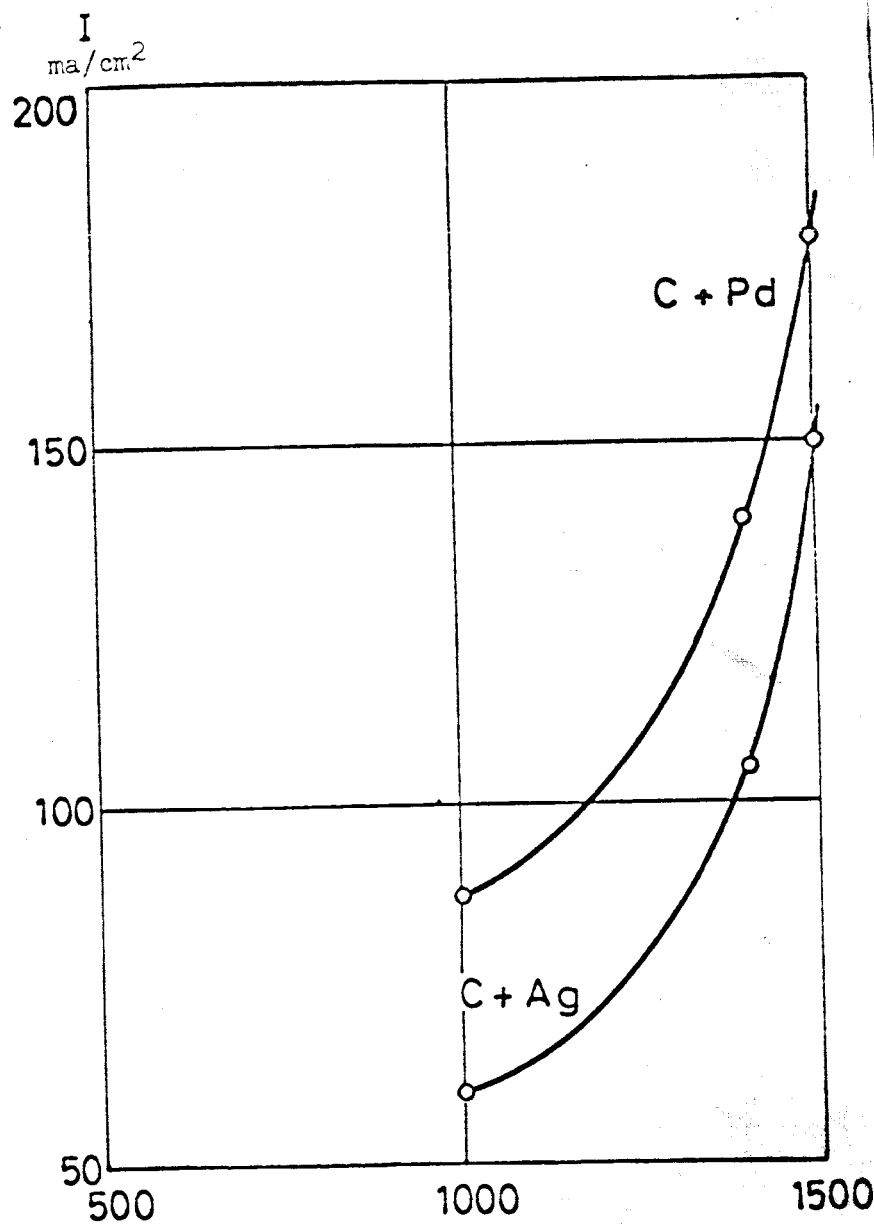


Fig.14 Influence of the Active Carbon Surface
on the Catalyst Activity

ties of the assembly (Bibl.64 - 65). These authors believe that only the chemical bonds are able to ensure high activity and that the simple mixtures have no function at all. So far as we are concerned, we have made an attempt to prepare catalytic mixtures on the base of carbon and metal powders (silver or palladium). The proportions by weight were the same as those obtained by chemical deposition, and the experiments were made with powders of differing granulometry (30 - 120 μ). A clear difference was observed between the behavior of the powder mixtures and that of the catalysts obtained by chemical deposition. With respect to the oxygen electrode, the currents collected on the mixtures were about three times weaker than those collected on the chemically treated catalysts. It should be mentioned, on the other hand, that the zero-current potential was positive and had a value of 200 mv for the mixture of carbon and powdered silver. In the case of electrodes on a palladium base, it was observed that the hydrogen was not adsorbed and that the equilibrium potential remained far from the reversible potential. Finally, it should be mentioned that the granulometry of the powders used has no influence on the behavior of the electrodes.

These statements seem to indicate that the surface activity of the metal-powder grains differs from that of the chemical depositions. It could be /27 imagined, in addition, that the carbon/palladium or the carbon/silver electrode actually constitutes a bielectrode, in which the carbon fixes a potential defined by the presence of residual oxygen in its lattice while the other metal fixes a potential defined for the gaseous reagent supplied to the electrode. The value of the total potential, resulting from the combination of these two voltages, depends on the relative surfaces of the two solids.

Thus, the idea of making use of metal powders must not be rejected. Such

a method is of considerable interest insofar as certain metals or compounds of relatively low cost but difficult to deposit by chemical means could thus satisfactorily replace products of high cost, such as noble metals.

How can the reactive zone in such electrodes be localized? It might be that the electrochemical reaction takes place at points where the microchannels abut the macrochannels. Thus, if the macrochannels are numerous, the entire surface of the macropores would be electrochemically active. Figure 13 shows the difference between the extent of the reactive zone in a pore of a conventional electrode and that in a macrochannel of active carbon. Any increase in the number of micropores would result in an increase of the active surface. The curve in Fig. 14 demonstrates clearly that the catalytic activity of an electrode increases considerably with the specific surface of the carbon used (about 90% of this surface corresponds to the microporosity).

Austin (Bibl.66), for the case of electrodes with very small pore diameter, considered a limitation of the gas feed by the Knudsen effect. However, it /28 is reasonable to imagine that the number of possible paths is so large that the quantity of gas, arriving at the reactive sites, is sufficient for preventing a restriction of the reaction, taking the catalytic activity of the solid into consideration.

It must also be stipulated that the binder must be sufficiently finely divided to be permeable to the gas so that, on the one hand, the microporous structure can be continuous and, on the other hand, the reactive sites will not be masked.

Thus, electrodes with gas feed for lateral diffusion exhibit the characteristics of a system in which the feed process of the reagents and the removal of the charges each depend on distinct geometric parameters. In fact, the dif-

fusion of the gas is dependent only on the number of microchannels present, whereas the evacuation of the charges is a function of the diameter of the large channels and of their number. The ohmic drop in the liquid phase is entirely negligible; conversely, the ohmic drop in the solid phase may be quite high since the contact resistance between the carbon and the metal collector is not zero.

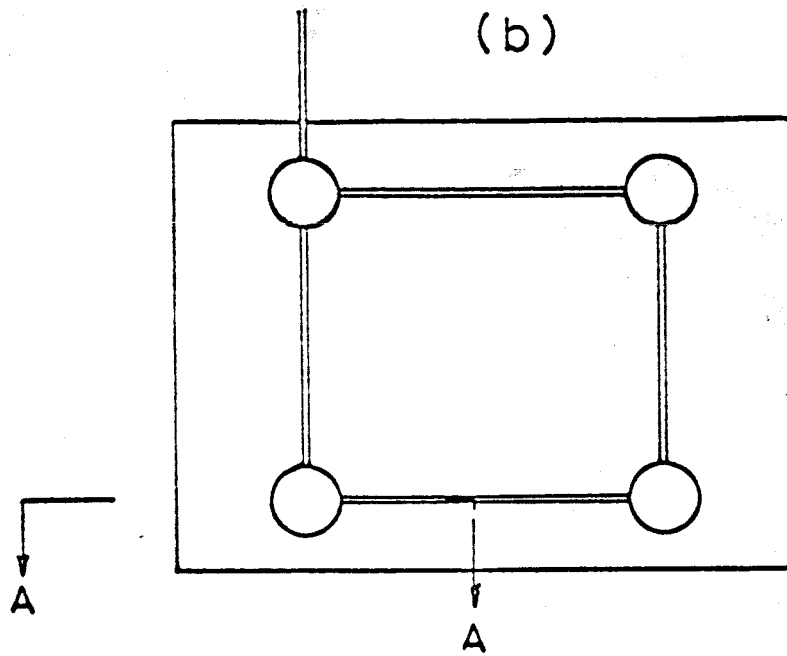
Electrodes of this type are specifically characterized by the fact that the surfaces, directly exposed to the gaseous flux, are negligible in size with respect to the total surface in contact with the electrolyte. It should be recalled that, in the classical porous electrodes, the surface washed by the gas is of the same order as the surface in contact with the electrolyte. For this reason, designers using such electrodes were always induced to use identical /29 structures. However, the possibility of localized feeding of reagent, together with the use of extremely thin electrodes, leads to the design of cells with promising potentiality of performance. The new structures, resulting from such electrodes, have the main advantage of being very compact and thus of ensuring high mass transport. The possible applications of this process have been covered in a patent application (Bibl.67).

2. Applications

In accordance with this localized feed principle of gaseous reagents, we designed cells that differ merely in the arrangement of the feed zone. Two systems were investigated:

a) Feed by "Studs"

In this system, the gas is supplied to the catalyst by means of rubber



(a)

Section A-A

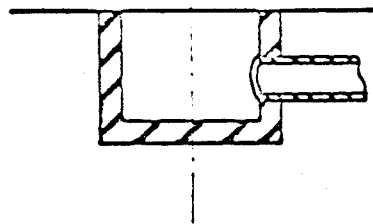


Fig.15

/29b

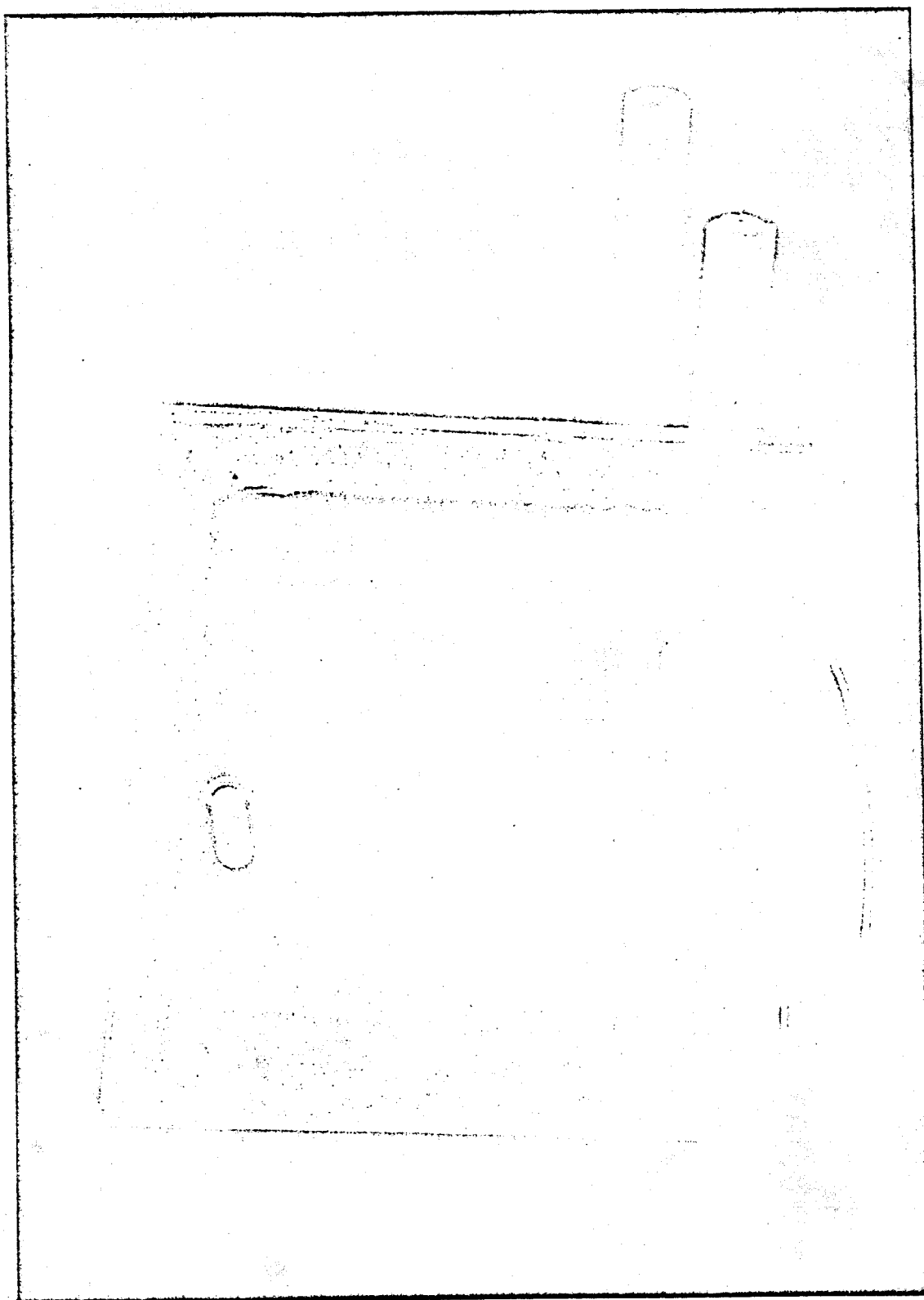


Fig.16

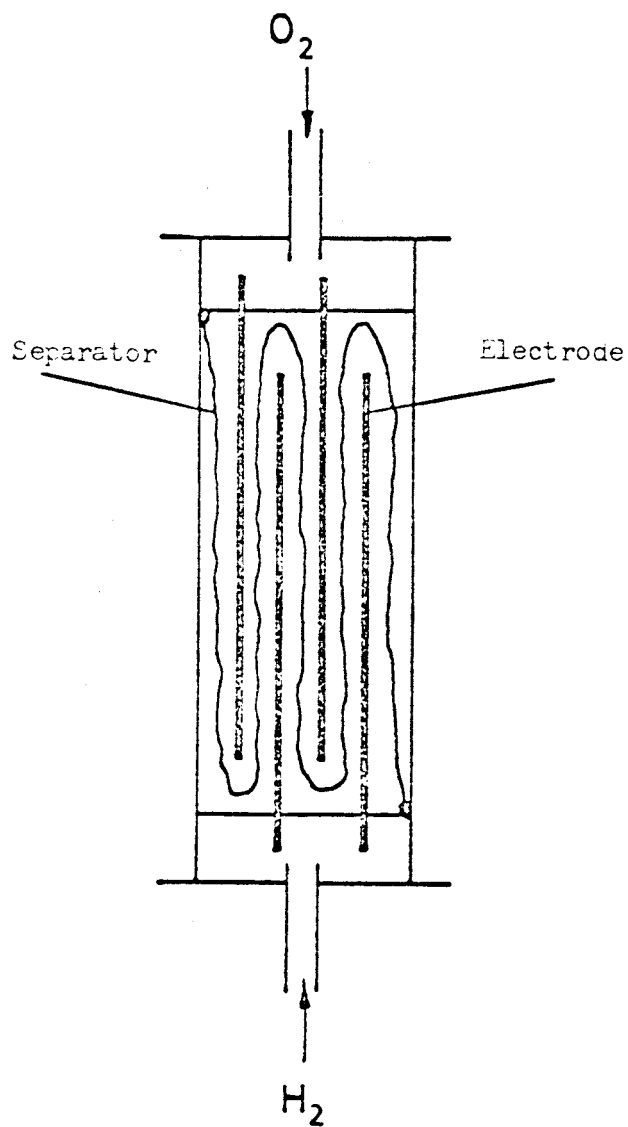


Fig.17 Schematic Sketch of a Fuel Cell Model
with Lateral Diffusion

studs or nipples (shown in Fig.15a) which are applied to the electrode under slight pressure. These studs are so arranged (Fig.15b) that the entire surface of the electrode can be supplied with reagent. The main drawback of this process is the difficulty of obtaining a sufficient seal between studs and carbon; this leads to a slight bubbling of the gas at this level. For this reason, the electrode and its feed unit are placed into a porous Nylon bag so as to avoid, as far as possible, any diffusion of the gas from one electrode to the other. This Nylon bag has the additional advantage of producing electric insulation of electrodes with opposite polarity.

b) Feed by Electrode Strip

/30

The rectangular electrodes are fed along two opposite strips. The latter are enclosed in a sort of liquid-proof bag into which the gas is introduced under a slight overpressure (several centimeters of water). The electrodes are separated by a porous Nylon partition, and the entire assembly is placed into two polyvinyl chloride flasks. It should be mentioned that, if the feeding surfaces are too close together, the amount of gas fed to the electrode will become too high to be entirely consumed by the electrochemical reaction; this will lead to a slight bubbling at the center portion of the electrode surface. The upper portion of the cell is provided with orifices for evacuating and recycling the gas. Small elements, based on this principle and having a power of approximately 1 watt, have been produced. A photograph of such a unit is shown in Fig.16.

A variant of this process consists in assembling several electrodes of the same polarity so as to form a type of comb. The edge of each electrode is hermetically sealed in a chamber containing the gaseous reagent under slight pres-

sure. The cell itself is obtained by grouping two units of inverse polarity, as shown in Fig.17. A strip of porous Nylon, slipped between the electrodes, prevents short-circuiting and, as in the above case, avoids diffusion of the excess gases.

This type of design is by far the most interesting with respect to compactness and mass transport of the elements. For this reason, this configuration was used for developing a prototype.

However, mounting of the electrodes presents some difficulty, with respect to pasting the electrodes into the gas feeder compartments. /31

Satisfactory seal at the level of the carbon, between the liquid and gas compartments, is ensured by slipping a silicone rubber collar around the electrode. To obtain a satisfactory seal, the paste must be sufficiently fluid at the instant of pouring, so as to slightly impregnate the carbon. However, the degree of fluidity must be accurately defined to avoid too deep a penetration into the catalyst. This would result in blocking the diffusion of gas into the carbon.

3. Behavior of the Electrodes

We studied the behavior of the electrodes before using them in a prototype. These electrodes consisted of rectangular nickel grids, coated with catalysts on an active carbon base. The thickness of the entire unit was at most 1 mm. The mounting was done so that the surface exposed to the gaseous atmosphere was about six times smaller than that in contact with the electrolyte.

It should be emphasized that, for reducing the weight of the electrodes, the metal grid can be replaced by a porous Nylon foil, coated with a thin layer of silver lacquer which makes it conductive. Numerous electrodes designed on

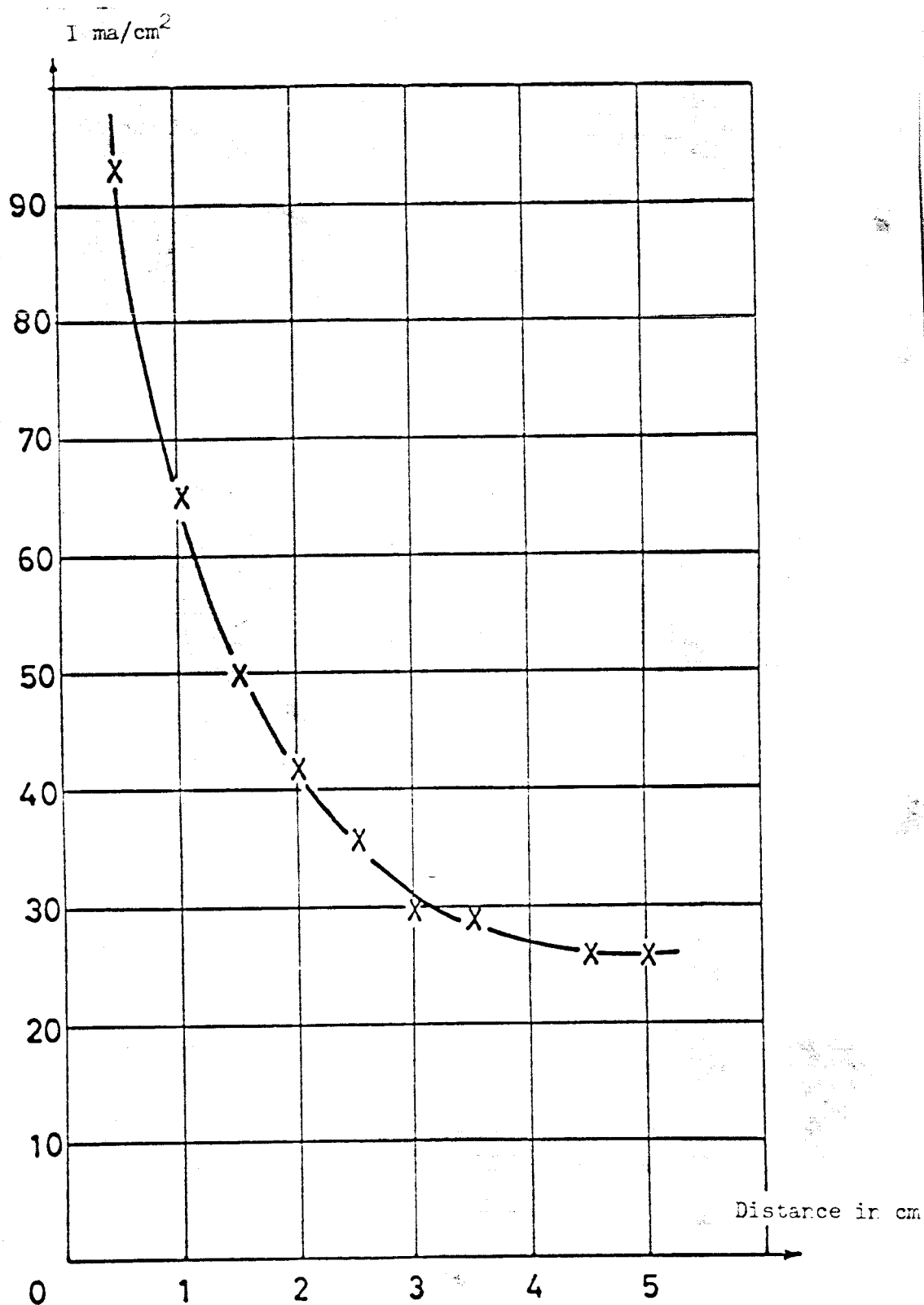


Fig.18 Density of the Collected Current as a Function of the Distance Covered by the Gas

this principle have been used in various experiments and were found entirely satisfactory.

To define the optimum dimensions of these electrodes, it is suggested to determine the mean free path of the gas in the catalyst, starting from the line of paste-up. For this, the mean density of the collected current was determined for electrodes of various dimensions. The curve in Fig.18 shows that, starting from a distance of about 2.5 cm, the rate of feed of the gas is no longer sufficient to ensure a noticeable increase in the collected current. Consequently, this particular dimension was adopted for all future work. The second dimension, corresponding to the length of the feed line, was fixed at 9 cm to facilitate the construction. Electrodes, laid out in this manner, were mounted in a half-cell to study their polarization as a function of the collected currents (Figs.19 and 20) and to investigate the influence of the partial oxygen pressure on this polarization (Fig.21). /32

During these experiments, we found that the catalysts used for oxidation of the hydrogen, in some cases, were highly sensitive to poisoning by certain gases. It seems that specifically sulfurous products, of which traces are sometimes present in commercial hydrogen, may become adsorbed on the electrodes and thus greatly impair their catalytic properties.

This poisoning is shown in the fact that the potential assumed by the electrode at equilibrium is located quite far from the reversible potential of hydrogen. This would confirm the hypothesis according to which a part of the active sites are occupied by the poison. It might also happen that the trichloroethylene used as solvent for the binder - if proper precautions are not taken when drying the electrodes - will remain in the carbon and thus limit the adsorption of the hydrogen.

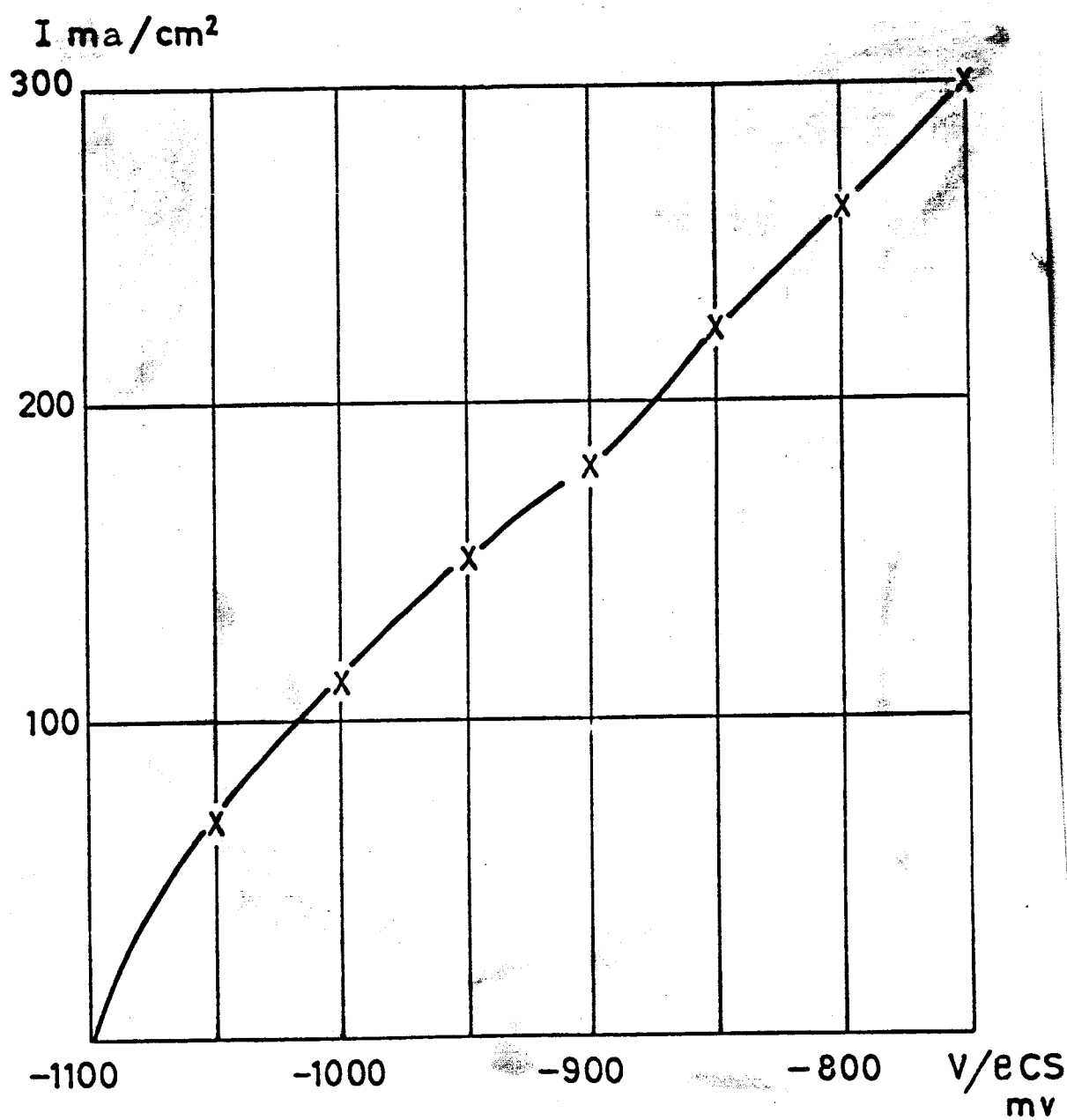


Fig.19 Polarization Curve of a Hydrogen Electrode

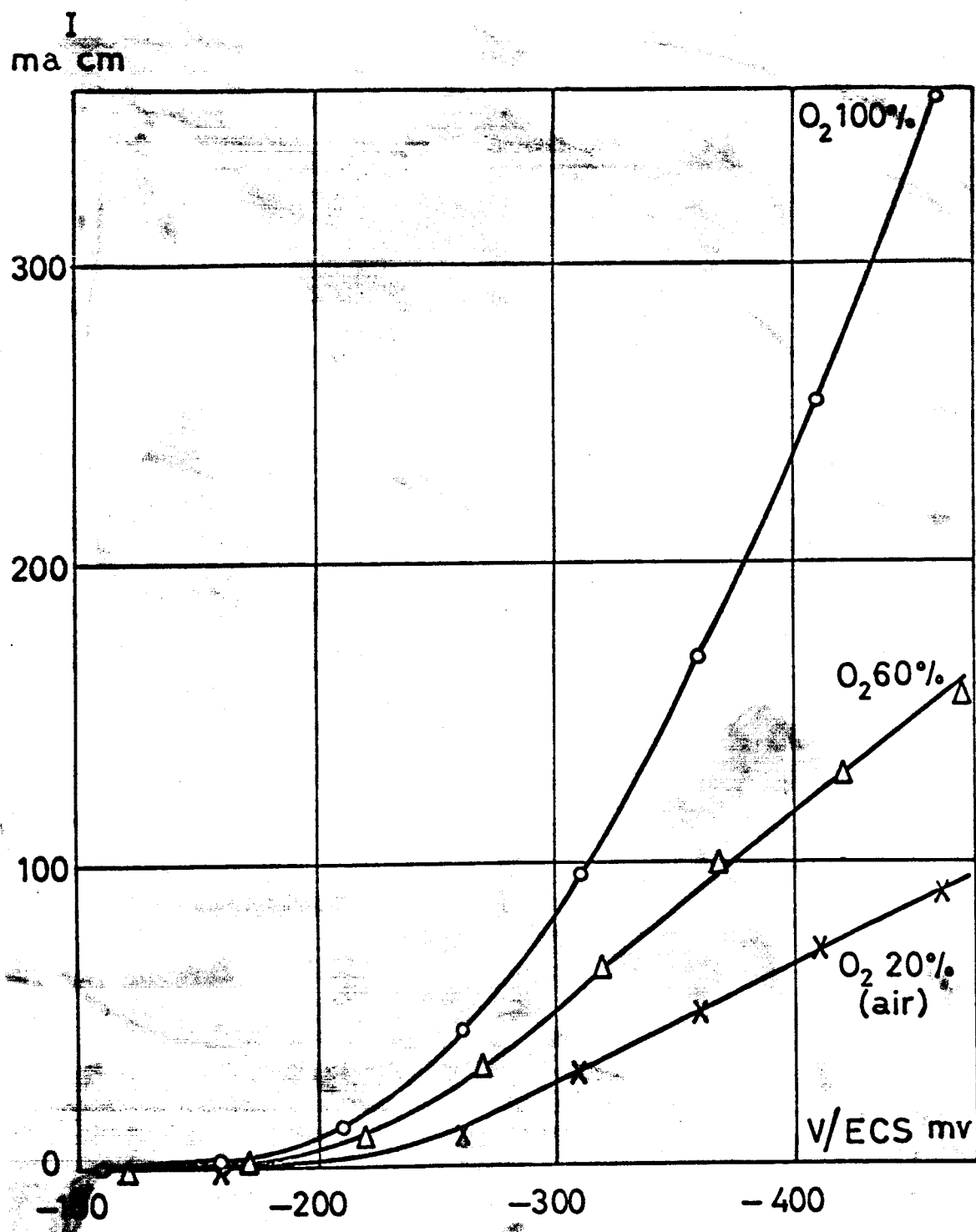


Fig.20 Influence of the Partial Oxygen Pressure on the Polarization Curves

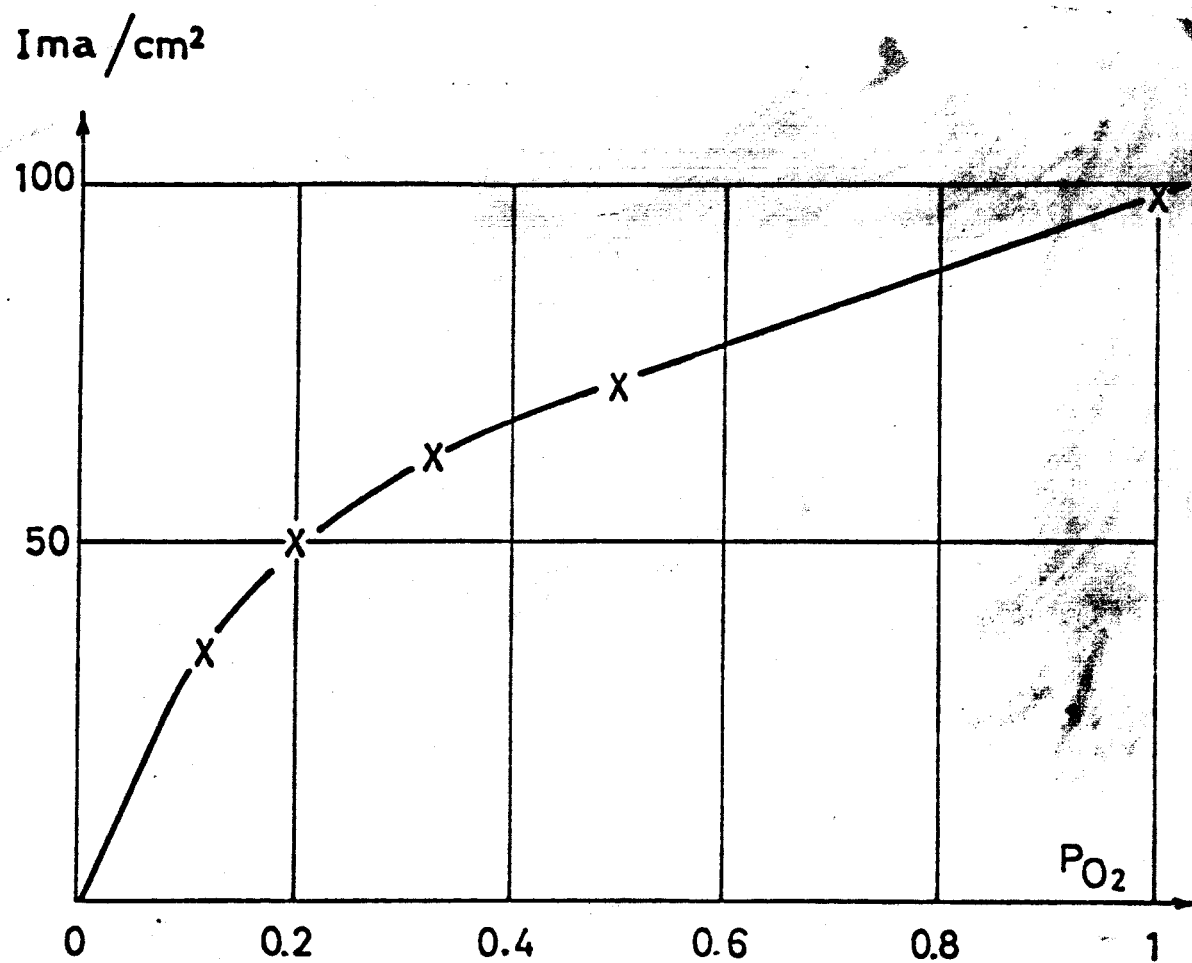


Fig.21 Influence of the Partial Oxygen Pressure on the Current

Finally, to complete the experiments on half-cells, we started a study of the variation in performance of these electrodes with time. Hydrogen electrodes and oxygen electrodes were placed under polarization, in a continuous test, for about 1000 hrs without resulting in a noticeable variation in current. These experiments are still in progress. /33

4. Development of Prototypes

A small one-watt element, operating on the principle of lateral diffusion, was used for studying the behavior of electrodes in the cell. The photograph in Fig.16 shows this particular element. The gas was fed from two strips opposite to the electrode. The current-voltage characteristic (Fig.22) and the power curve (Fig.23) were plotted for this particular cell. We also attempted to define the influence of the temperature on the depolarization of the electrodes. The curve plotted in Fig.24 indicates that the effect of the temperature is rather extensive.

In fact, for an increase in temperature from 25 to 60°C, the collected current increases by about a factor of two. It is obvious that an assembly into cells of several such modules would be conceivable. We used this principle to design a generator of a power of several tens of watts (power varying with the operating temperature, which might fluctuate between 20° and 80°C).

The characteristics of this generator are given in Appendix I.

5. Conclusions

/34

In the work reported here, after a brief analysis of the basic operating characteristics of a conventional electrode for fuel cells, an attempt was made to define systems for which the feed conditions of the reagent and the evacua-

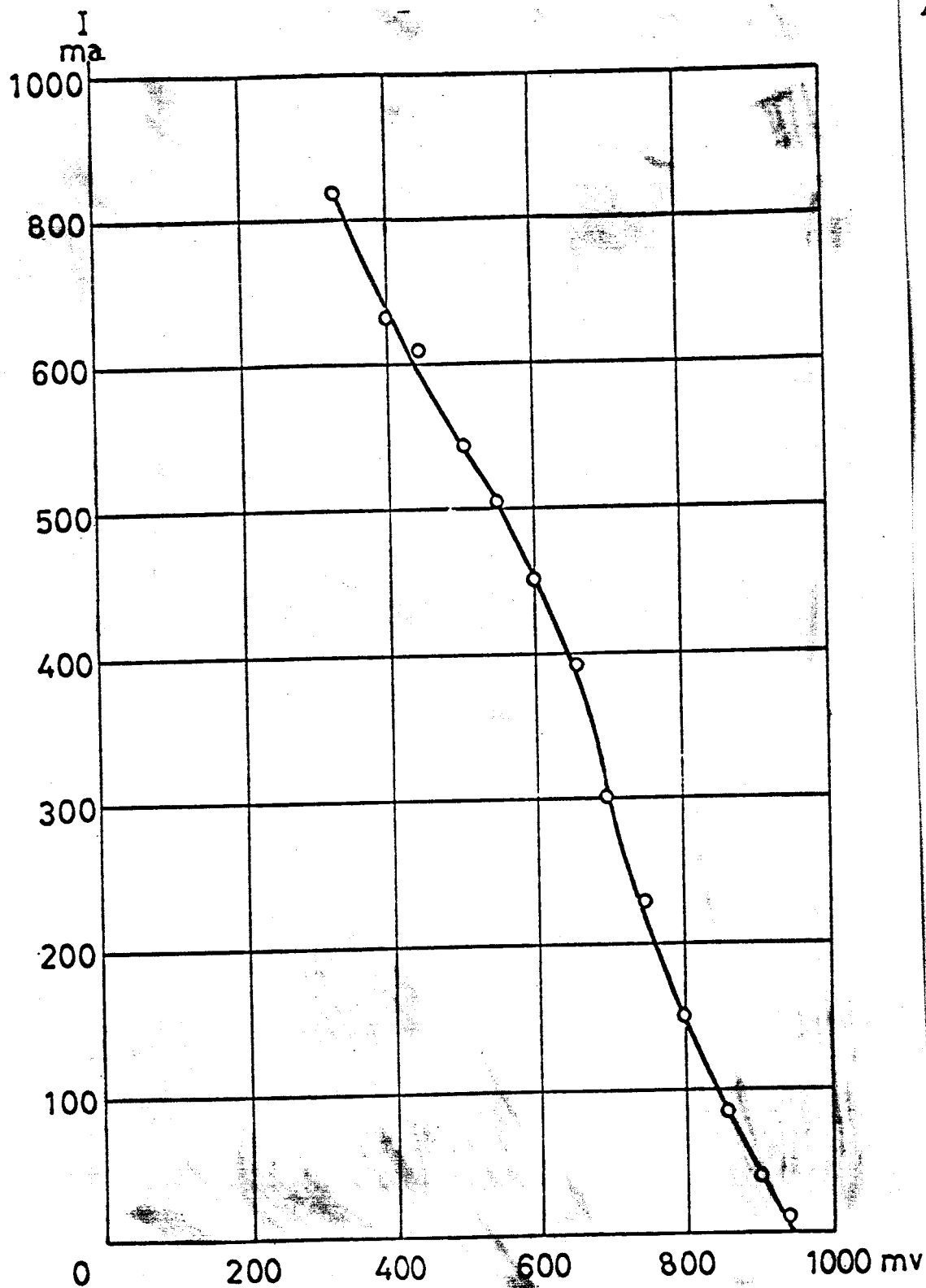


Fig.22 Polarization Curve of a Lateral Diffusion Cell

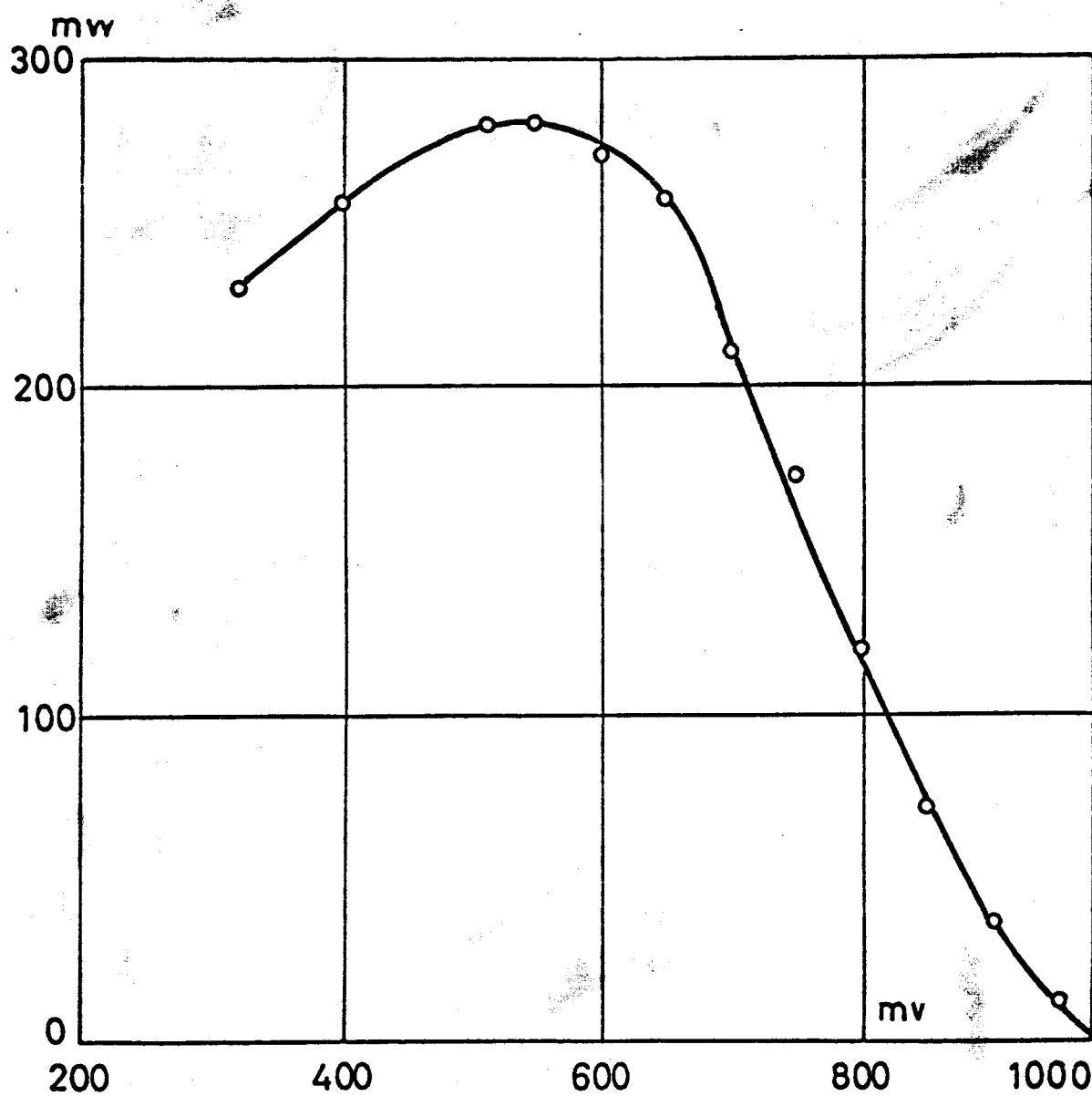


Fig.23 Power Curve of a Lateral Diffusion Cell

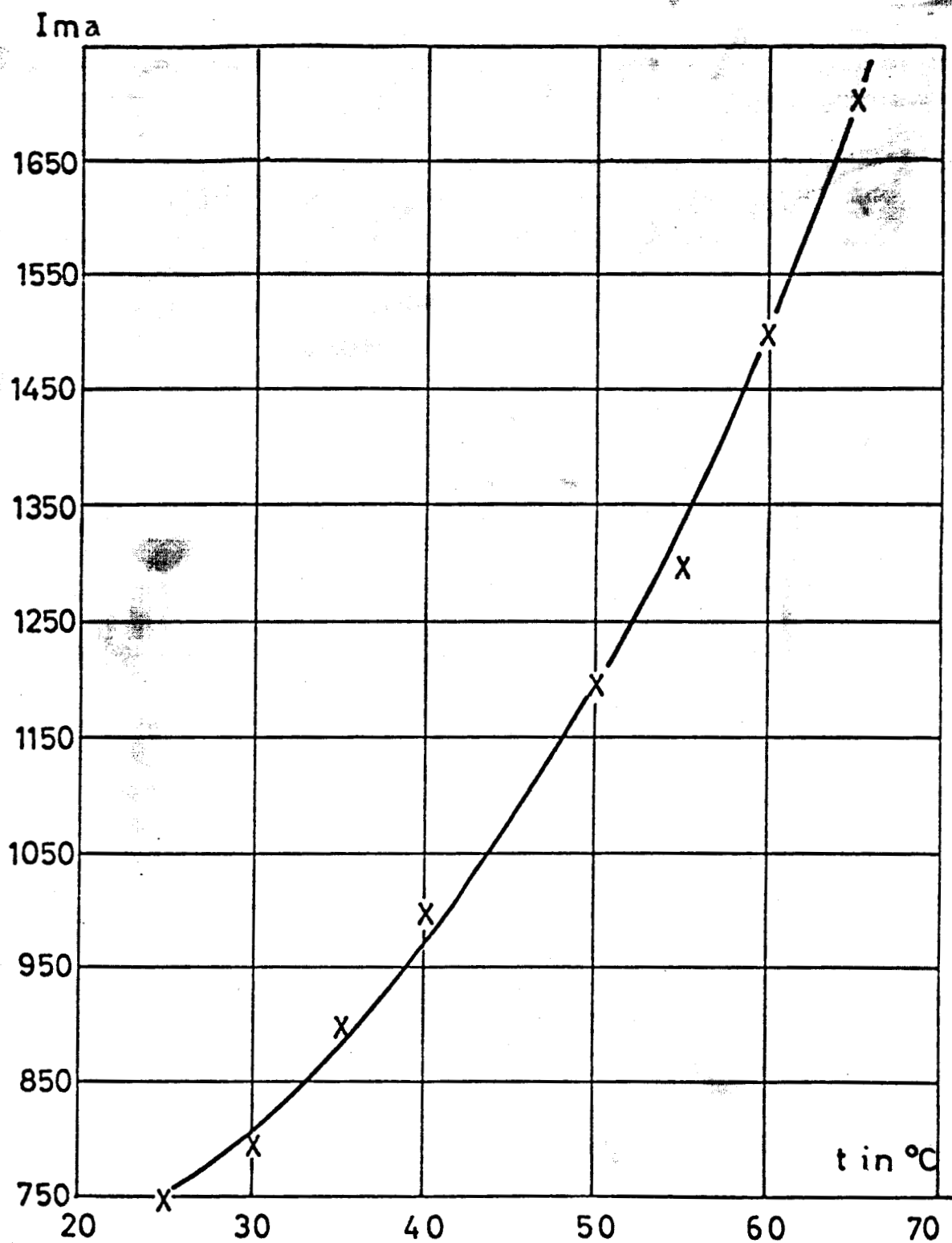


Fig.24 Influence of Temperature on the Depolarization

tion of the charges no longer were antagonistic, so as to obtain conditions under which the reaction of the electrode would become much more efficient. Among others, we investigated rotary half-immersed electrodes for which we were able to demonstrate a decoupling process of gas feed and electrochemical consumption.

In experiments with such electrodes, we particularly studied the operating behavior of catalysts on an active carbon base. This led to a special mode of feeding the gaseous reagent to the reactive sites, for this particular type of catalyst.

The basic characteristic of this mass transport is connected with the twin, microporous and macroporous, structure of active carbons. Thus, we determined that the smallest pore network basically was used for transporting the gas while the largest pores, filled with electrolyte, represented the electrochemically active surface. Since the density of the micropores is quite high, the gaseous flux will be sufficient for properly feeding the entire surface of /35 the macrochannels, even after covering paths several centimeter in length. Consequently, these electrodes are characterized by an excellent gas feed over the entire area of the solid-liquid interface. Under these conditions, the conventional mode of feeding electrodes, in which the surfaces in contact with the gas are of the same order as the surfaces washed by the liquid, must be considered obsolete. In fact, we were able to demonstrate that the electrodes can be fed with gas from dry regions, whose total surface can thus be greatly reduced. This layout, which differs greatly from standard types, permits the construction of fuel cells whose principal characteristic is a high mass transport. This means that designs generally used for batteries and standard electric cells, so as to obtain maximum compactness, can now be used in the construction

of fuel cells.

The results obtained on modules of several watts, permit a prediction, with rather reliable estimate bases, of the possible mass transport output of such generators. If every possible effort is made to reduce the weight of the prototype, if the operating temperature is raised to about 80° by self-heating, if the interelectrode spacing is reduced by using grids for current collection weighing about half of the grids used in our preliminary designs, and - finally - if the casings are manufactured of lower-density materials and thus will have less thickness, the weight of a one-kilowatt cell could be reduced to about 5 kg.

By using an electrode arrangement, tested in several experiments and /36 consisting of a spiral arrangement of the electrodes, it seems possible to obtain a mass transport, under the above operating conditions, of about 3 kg/kw. To give a general idea, it should be mentioned that fuel cells in use either in France or in the United States, have mass powers of the order of 60 kg/kw. Research centers in the US believe that they will be able to produce, within a relatively short time, hydrox fuel cells of a weight of 20 kg per kilowatt.

Another interesting property of this process is its low original cost, due to the simplicity of design and materials of conventional type. The casings for the cells can be manufactured by molding or sandwiching of various plastic laminates (polyvinyl chloride, Teflon, etc.). With respect to fabrication of the electrodes, processes of these types would eliminate the need for presswork and fritting which are delicate and tedious techniques; in addition, it would be possible to get along with very small quantities of catalysts, as mentioned above. Recent experiments also have demonstrated that it is of no importance what type catalyst is used together with the carbon. Thus, the process is

given an entirely general character.

Finally, it should be mentioned that the operating conditions (low pressure and low temperature) make it possible to use air as oxidizer, at satisfactory yield. In fact, since the electrodes operate at very low pressures not exceeding a few centimeters of water, it is no longer necessary to compress the gas, which naturally constitutes a considerable saving.

The extremely interesting results obtained for hydrox systems seem to /37 indicate that this process can be advantageously used in cases of cells that permit a direct oxidation of hydrocarbons.

In conclusion, we would like to mention that the attempts made to understand the operating mechanism of gas electrodes have led to the development of electrochemical generators whose performance, although still open to further improvement, is at least equal to that obtained with classical procedures. Because of this fact, the application possibilities of fuel cells have been greatly extended and it seems quite likely that, in view of the fact that similar efforts are made on the study of catalysts, application such as propulsion of spacecraft no longer is in the domain of Utopia.

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Written Examination for Master of Engineering

Specialty: Electrochemistry

NATIONAL CONSERVATORY OF ARTS AND TRADES

Denis Doniat

Subject: Contribution to the Study of New Designs
for Fuel Cells

APPENDIX I

GENERATOR WITH LATERAL DIFFUSION ELECTRODES

Paris 1965

1. Characteristics of the Elementary Module

/1

Number of electrodes: Eleven, 5 of which are anodes and 6 cathodes.

Type of electrodes:

Cathodes: active silvered carbon, on a nickel grid;

Anodes: palladium-coated active carbon, on a nickel grid.

Thickness of electrodes: about 0.6 mm.

Weight of electrodes: about 30 gm.

Weight of casing: about 30 gm.

Weight of electrolyte: about 50 gm.

Total weight of module: 110 gm.

Maximum power: 2.5 w at 25°C,

5.5 w at 55°C.

Operating pressure for oxygen and hydrogen: about 20 gm/cm².

Mass transport at 55°C: 20 kg/kw.

2. Optimization of Mass Transport

a) Reduction in Total Weight of the Module

The weight of the presented module is distributed as follows:

27% for the casing,

46% for the electrolyte,

27% for the electrodes.

A considerable gain could be obtained by the following measures:

/2

1) Casing: the gain here could be 50%.

a) The elementary module of a 1-kw generator would have more electrodes than the presented module. This would reduce the weight of the casing for a

module of larger dimensions.

b) The materials used could be about 30 - 50% less in thickness than at present. In addition, materials of lower density can be selected.

c) The lead-in boxes for the gas can be considerably reduced in dimensions.

d) Manufacturing the casings on an industrial scale would reduce the amount of material required.

2) Electrodes: For these, the gain could reach 40%.

Current-collecting grids of less thickness could be used, or the collector holder could be made of metallized plastic.

The thickness of the carbon can be greatly reduced by improving the sealing units at the level of the gas/electrolyte box.

3) Electrolyte: The gain here could be 70%. The saving is one of the largest possible within the unit.

The volume of electrolyte could be reduced by improving the mutual pasting ratio of the electrodes.

The volume of electrolyte could be reduced by increasing the compactness 13 of the electrodes, which could be designed in the form of spirals.

b) Increase in Power of the Module

1) The power could be increased by improving the efficiency of the catalysts used. It is obvious that optimum results have not yet been reached. It is suggested to make further studies on the type of catalyst and its degree of dispersion; in addition, carbons of a larger specific surface could be used.

2) The cell could be operated at a higher temperature (70 - 90°C) which would increase the output by a factor of 2 to 3, as demonstrated by tests on half-cells. It should be mentioned that this temperature can be obtained readi-

ly by self-heating.

c) Conclusions

The above estimates indicate that the mass transfer of a hydrogen-oxygen generator, using "lateral diffusion" electrodes, may reach values of the order of 5 kg/kw, but at the expense of some lowering in perfection.

In addition to the mass transfer increase which constitutes the principal characteristic of this type of generator, the following properties should be mentioned: /4

1) Relatively low cost price for the electrodes: production by sputtering rather than by pressing or fritting. The main catalytic material will still be carbon. Low cost price of the casing (conventional plastic materials) and assembly compatible with production in series.

2) Utilization of gases at low pressure. Thus, in the case of using air as the oxidizer, the power lost by compression of the gas, before its intake, is practically compensated.

3) Since the "ohmic" term is negligible in the mentioned polarization, it is possible to use - without noticeable drop in performance - less conducting electrodes than KOH_2N , a point that might be of interest when other fuels are used (hydrocarbons).

4) The operating principle of the electrodes (carbon used as vehicle for the gas and adsorption on the catalytic element which is either deposited on or incorporated into the electrode) permits the utilization of widely differing catalysts, which cannot be used in molded and fritted electrodes.